

RL-TR-96-264
Final Technical Report
April 1997



WAVEGUIDE AMPLIFIERS USING Cr:FORSTERITE AND Cr:YAG

Cornell University

Clifford Pollock

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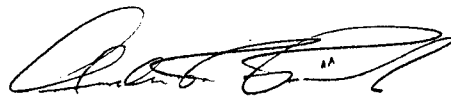
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APPROVED:



ANDREW R. PIRICH
Project Engineer

FOR THE COMMANDER:



DONALD W. HANSON, Director
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REPORT DOCUMENTATION PAGE			Form Approved OMB No. 0704-0188	
Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.				
1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE April 1997	3. REPORT TYPE AND DATES COVERED FINAL, May 95 - May 96	
4. TITLE AND SUBTITLE WAVEGUIDE AMPLIFIERS USING Cr:FORSTERITE AND Cr:YAG			5. FUNDING NUMBERS C - F30602-95-C-0103 PE - 62702F PR - 4600 TA - P4 WU - PS	
6. AUTHOR(S) Clifford Pollock				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Cornell University Office of Sponsored Programs 120 Day Hall Ithaca NY 14853			8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) Rome Laboratory/OCPA 25 Electronic Pky Rome NY 13441-4515			10. SPONSORING / MONITORING AGENCY REPORT NUMBER RL-TR-96-264	
11. SUPPLEMENTARY NOTES Rome Laboratory Project Engineer: Andrew R. Pirich, OCPA, (315) 330-4147				
12a. DISTRIBUTION AVAILABILITY STATEMENT Approved for Public Release; Distribution Unlimited			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) Using an optical composite of Cr-doped forsterite nanocrystals embedded in a polymer host with matching refractive index, a waveguide has been built which displays optical gain up to 3dB/cm at 1.25 μ m. This gain magnitude is significantly larger than that obtained from Er-doped glass fibers, and illustrates the potential for optical composite materials. In a similar experiment, we demonstrated optical gain from Cr:diopside at 800 nm. Diopside has never been grown in large single crystal form, so this is the first report of stimulated emission from this material. We demonstrated stable femtosecond pulse generation from a Cr:YAG laser using a Saturable Bragg Reflector as an end mirror of the laser cavity. This technique of modelocking allows long term femtosecond laser operation of the laser with reasonable (50 mW) output power and tuning from 1488 nm to 1535 nm. In a similar experiment, the Saturable Bragg Reflector was put into a color center laser cavity and generated stable modelocked pulses from this laser. Pulse duration extended from 100 femtosecond up to 2 psec depending on the focussing of the intracavity beam on the Saturable Bragg Reflector.				
14. SUBJECT TERMS femtosecond pulse generation, thin film composite amplifier, Cr:diopside crystal, thin film waveguides			15. NUMBER OF PAGES 20	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UNLIMITED	

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Waveguide Amplifiers Using Cr:Forsterite and Cr:YAG

I. Abstract

Using an optical composite of Cr-doped forsterite nanocrystals embedded in a polymer host with matching refractive index, a waveguide has been built which displays optical gain up to 3 dB/cm at 1.25 μm . This gain magnitude is significantly larger than that obtained from Er-doped glass fibers, and illustrates the potential for optical composite materials. In a similar experiment, we demonstrated optical gain from Cr:diopside at 800 nm. Diopside has never been grown in large single crystal form, so this is the first report of simulated emission from this material.

We demonstrated stable femtosecond pulse generation from a Cr:YAG laser using a Saturable Bragg Reflector as an end mirror of the laser cavity. This technique of modelocking allows long term femtosecond laser operation of the laser with reasonable (50 mW) output power and tuning from 1488 nm to 1535 nm. In a similar experiment, the Saturable Bragg Reflector was sput into a color center laser cavity and generated stable modelocked pulses from this laser. Pulse duration extended from 100 femtosecond up to 2 psec depending on the focussing of the intracavity beam on the Saturable Bragg Reflector.

II. Introduction

The thrust of this research effort has been to create thin film waveguides based on Cr:Forsterite or Cr:YAG, and to generate ultrafast pulses using these lasers. In single crystal form these materials provide broadly tunable laser emission over the 1.2 -1.3 μm and 1.35-1.6 μm regions of the near infrared spectrum, respectively. Typical power and tuning ranges for the Cr:Forsterite and Cr:YAG lasers are shown in Figure 1. The tuning range of these lasers overlaps the popular communications bands, which makes the materials interesting not only as sources for optical communication system testing, but also as potential optical amplifiers. In this report we describe the development of the first waveguide amplifiers based on a novel composite material of Cr:Forsterite nanocrystals embedded in a polymer host. Gains as high as 3 dB/cm were observed.

A significant application of these lasers is in the generation and application of femtosecond pulses. To date these lasers have been modelocked using Kerr Lens Modelocking (KLM) techniques. Unfortunately the KLM technique is extremely sensitive to perturbations, and does not deliver stable pulse trains with either the Cr:YAG or Cr:forsterite lasers. After much failed effort at creating stable KLM operation of the laser, we adapted the use of saturable absorbers in the cavity. Using a technique pioneered by Wayne Knox at Bell Laboratories, we were able to generate a very stable pulsetrain of femtosecond pulses from

the Cr:YAG laser. The method is quite universal: we have used the same device to modelock a color center laser with stable femtosecond operation. In this report we describe the saturable absorber used to modelock these lasers, and present preliminary results.

III. Thin Film Composite Amplifiers

We have developed ways to create thin film structures of Cr:Forsterite and Cr:YAG laser materials which can be used to make waveguide amplifiers and laser sources. Thin film structures offer several advantages: 1) they require much less material so that the laser development is not limited by single crystal growth, 2) the optical confinement of a waveguide allows the use of high intensities over long ranges in the gain medium, promoting more efficient energy extraction and allowing for pumping from non-ideal optical sources such as laser diodes, and 3) the dimensions of the waveguide are compatible with direct fiber optic input and exit. These properties, when taken together, allow for the possibility of creating small, integrable optical devices such as sources and amplifiers, which can be incorporated into optical systems.

There has been significant previous work in waveguide lasers. The first demonstration was perhaps that of Koester and Snitzer[1] in 1964, where they demonstrated a Nd-doped fiber laser with gain exceeding 5×10^4 in a one meter length. More recently, there has been a great deal of work in rare-earth doped fiber lasers, led primarily by the efforts of Payne and Hanna in Southampton, United Kingdom. Among their advances, they invented the Er-fiber amplifier[2], and have demonstrated numerous other fiber and waveguide lasers based on rare-earth dopants in glass hosts[3]. Except for the well known Er fiber amplifier, most of these devices operate on one or two single lines, and are not tunable. Erbium allows a small tuning range around $1.55 \mu\text{m}$ (about 30 nm), which has made it useful for certain optical communication system applications. There has been a great deal of activity in search of an amplifier for the $1.3 \mu\text{m}$ region, however to date there has not appeared a system which rivals Er-doped fibers in performance at this shorter wavelength. Very little work has been done using transition metal ions to make amplifiers in waveguides. We have explored in this work the use of Cr as a suitable dopant for waveguide amplifiers. The Cr ion has a significantly broader emission band, and could display gain over a large spectral region in the near infrared.

Unlike most of the rare-earth doped fibers which are based on glass, transition metal ions generally must be hosted in a crystalline environment. This is due to the nature of the optical emission from these ions. In rare-earths, the optically active electrons generally belong to the *f*-shell, and are not involved in bonding to adjacent ions in the host. Therefore the optical transitions are relatively independent of the host. With transition metal ions, the optical transitions occur with the *d*-shell electrons, which are directly involved in the covalent bonding to adjacent ions in the host. Therefore the optical properties of the ion are critically determined by the host lattice. Amorphous lattices such as glass fibers have not proved to be good hosts for Cr-ions, so Cr-lasers must be developed in crystalline materials. This adds a new dimension of difficulty to the development of thin film waveguide lasers based on Cr-doped materials.

It is very difficult to grow an epitaxial layer of a crystalline material on a substrate that does not perfectly lattice match to the crystalline film. This limitation is mostly due

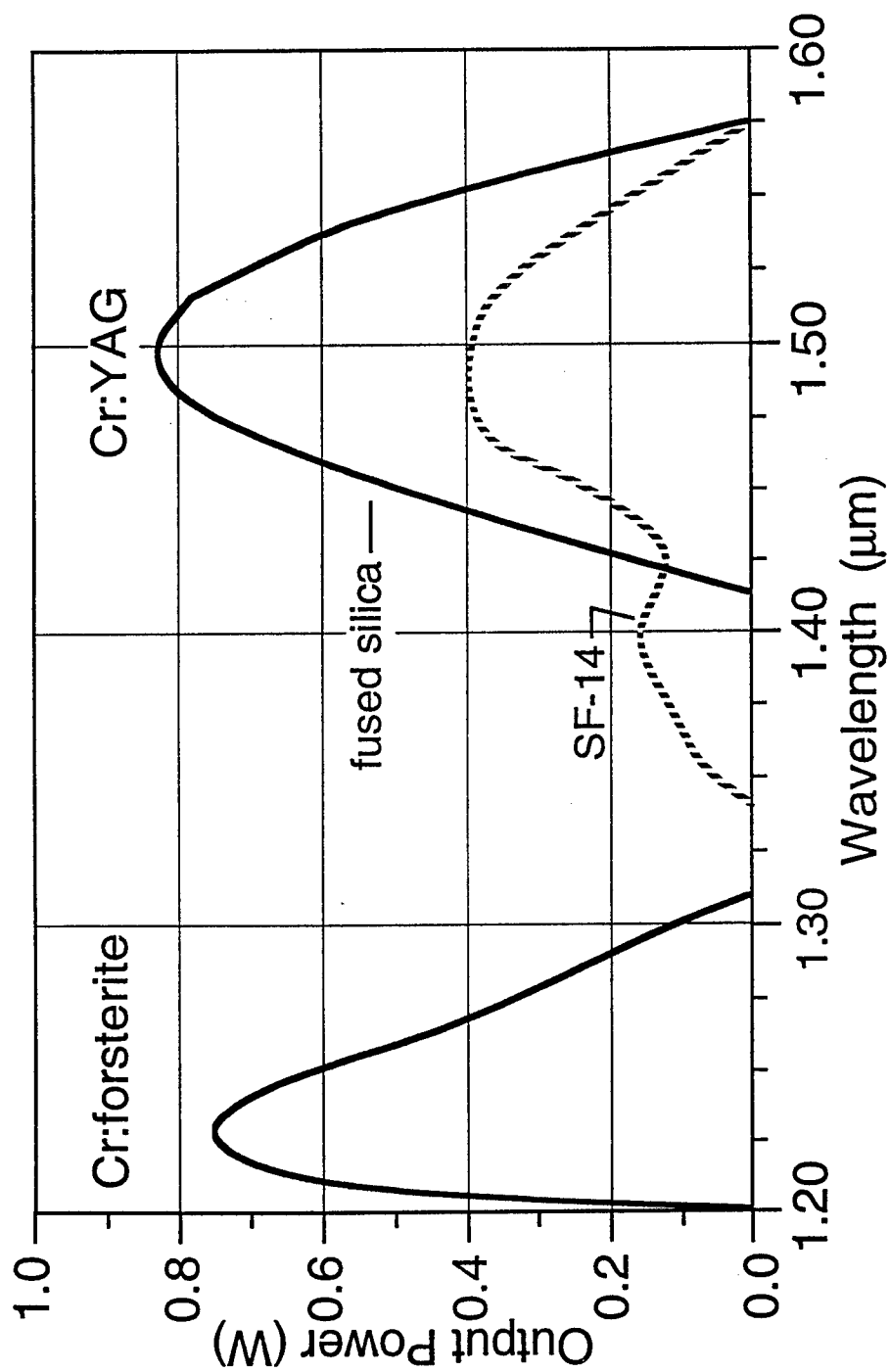


Fig. 1. Tuning curves for the Cr:YAG and Cr:Forsterite Lasers.

to strain that occurs at the interface between the substrate and epitaxial layer. Our initial interest in creating a thin film amplifier of Cr:Forsterite immediately ruled out the possibility of growing epitaxial layers due to the difficulty in finding a substrate which had the same crystal structure and dimensions of forsterite, and which had the lower refractive index necessary for creating an optical waveguide. Attempts to grow Cr:forsterite on amorphous substrates such as glass or silica, or on non-lattice matched substrates such as CaF_2 led to polycrystalline films which displayed strong scattering.

We were quickly led into considering a novel type of material based on composites of nanocrystals suspended in a refractive index matched host. With the help of Prof. Chris Ober in the Cornell Material Science and Engineering Department, we suspended a powder of Cr-doped forsterite particles in a refractive-index matched polymer, and created what we call an optical composite.

By making small particles of a solid state laser material and embedding them in a refractive index matched polymer matrix, the normal crystal growth procedure needed to produce laser quality single crystals is bypassed. This dramatically increasing the number of solid state materials that can be used as laser gain media. Further, the optical composite material can be made into thin film waveguides, allowing for long interaction lengths useful for low gain systems, optical amplifiers, and distributed integrated optical applications.

To minimize optical scattering, the refractive index of the particle and host need to be identical. This is not always possible as some optically useful crystals, such as forsterite, are uniaxial or biaxial. Therefore, we can expect that there may be scattering losses associated with the randomly oriented particles in a index-matched host medium. The Rayleigh scattering due to these particles (assuming the particle density is below the percolation point) is given by

$$\frac{P_{\text{scat}}}{P_0} = \rho \frac{(n' - n)^2}{n^2} \frac{V^2}{\lambda^4} 24\pi^4$$

where ρ is the number of nanocrystals per unit volume, n' is the index of refraction of the scatterers, n is the index of refraction of the host material, V is the volume of a single scatterer, and λ is the wavelength of light. It is desirable to make the particle size as small as possible, since scattering is proportional to the fourth power of the particle radius. Our calculations indicate that for particle diameters below 100 nm, scattering becomes insignificant compared to the achievable gain in most laser materials.

We constructed an optical composite by synthesizing nanocrystals of Cr-doped forsterite ($\text{Cr:Mg}_2\text{SiO}_4$), and embedding the crystals in a polymeric matrix with a matching refractive index. The optical composite film was constructed by dispersing 10 wt% Cr:forsterite nanocrystals in a copolymer made from tribromostyrene and naphthyl methacrylate which had a refractive index of $n = 1.65$ to match that of the forsterite particles. The mixture was ultrasonicated until the particles were dispersed, and then puddle cast on a level glass substrate. After drying slowly overnight the film was annealed at 200°C for 2 hours to remove excess solvent and to relieve strain in the film. This method resulted in films approximately $3 \mu\text{m}$ thick and 2-3 cm long. The ends of the structure were cleaved to make faces for end-fire coupling of the pump and probe signals in the amplification experiment.

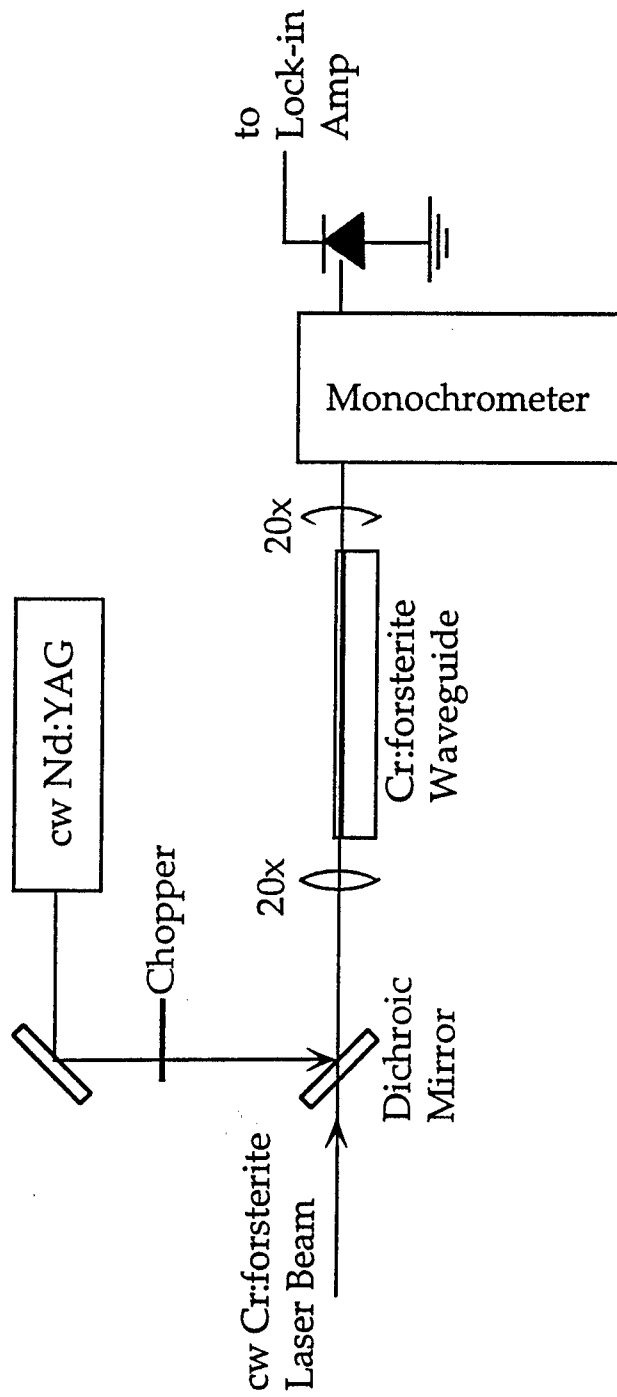


Fig. 2. Schematic configuration of waveguide gain measurement.

Using an optical setup as shown below in Figure 2, the waveguide was optically pumped with a chopped Nd:YAG laser operating at $1.06\ \mu\text{m}$, and probed with a cw beam at $1.25\ \mu\text{m}$ obtained from a cw forsterite laser. The beams were coupled onto and off of the waveguide using 20x microscope objectives. There was no attempt to create lateral confinement in this structure. The waveguide was approximately $2\ \mu\text{m}$ thick.

The composite waveguide was 1 cm in length, and displayed a maximum small signal (differential) gain of 3 dB, as shown in Fig. 3. To put this result in perspective, the widely used Er-doped fiber amplifier displays gains of approximately 3 dB/m (at $1.53\ \mu\text{m}$), or about 1% of the gain of the Cr:forsterite composite waveguide. Of further significance, the Cr:forsterite gain exceeds the best reported gains obtained from single crystal Cr:forsterite samples [4]. We believe the reason that the composite has a higher gain than a pure single crystal is a result of the high Cr-concentration in the forsterite nanocrystals. The low temperature chemical synthesis process used to create the nanocrystals achieves a higher concentration of Cr^{4+} ions in the proper tetrahedral lattice site than occurs in the traditional high temperature Czochralski growth from a seeded melt.[3]

There are still several aspects of this new device that need further investigation. One concern with the composite structure is the reduced heat conduction that the nanoparticles will have compared to that of single crystals. The single crystal forsterite laser is severely sensitive to thermal loading, primarily due to increased non-radiative relaxation of the excited state. We expect the nanoparticles to display similar characteristics; in fact due to the poorer thermal conductivity of the polymer, we expect that the thermal effects in the nanocrystal composites will be worse. To date we have observed no thermal loading on the performance of the composite waveguide, perhaps indicating that the thin ($\approx 2\ \mu\text{m}$) polymer allows sufficient heat to pass into the substrate before the temperature can build up to a significant level.

A second parameter of the amplifier that we still need to measure is that of gain saturation. We do not know the saturation intensity of this device.

A final problem that we are presently investigating concerns the condition of the end face on the optical waveguides. The reported 3 dB of gain was a differential gain, not a net gain. There is significant coupling loss of the input beam due to the poor condition of the end of the polymer waveguide. As a result, the input and pump beam suffered significant loss in coupling on and off the waveguide.

We spent a significant amount of time exploring methods to cleave a polymer-coating on a glass substrate. Attempts to simply cleave the glass substrate (a microscope objective approximately 2 mm thick) led to the formation of ragged edges on the polymer, even when the glass was cleaved straight. Apparently the bonding strength of the polymer to the glass is less than the bonding strength between adjacent polymer molecules. The next process we attempted was to cool the waveguides, freezing the polymer waveguide. We hoped this would enhance its tendency to cleave. Unfortunately, when the temperature of the glass-polymer waveguide was brought down to -100°C , the polymer coating peeled off of the glass substrate. Apparently the expansion coefficients of the two materials are different. We have considered finding a different substrate with a better matched expansion coefficient, but at this time we do not have a good value for the expansion coefficient of the polymer host. We are working with Prof. Chris Ober of the Cornell

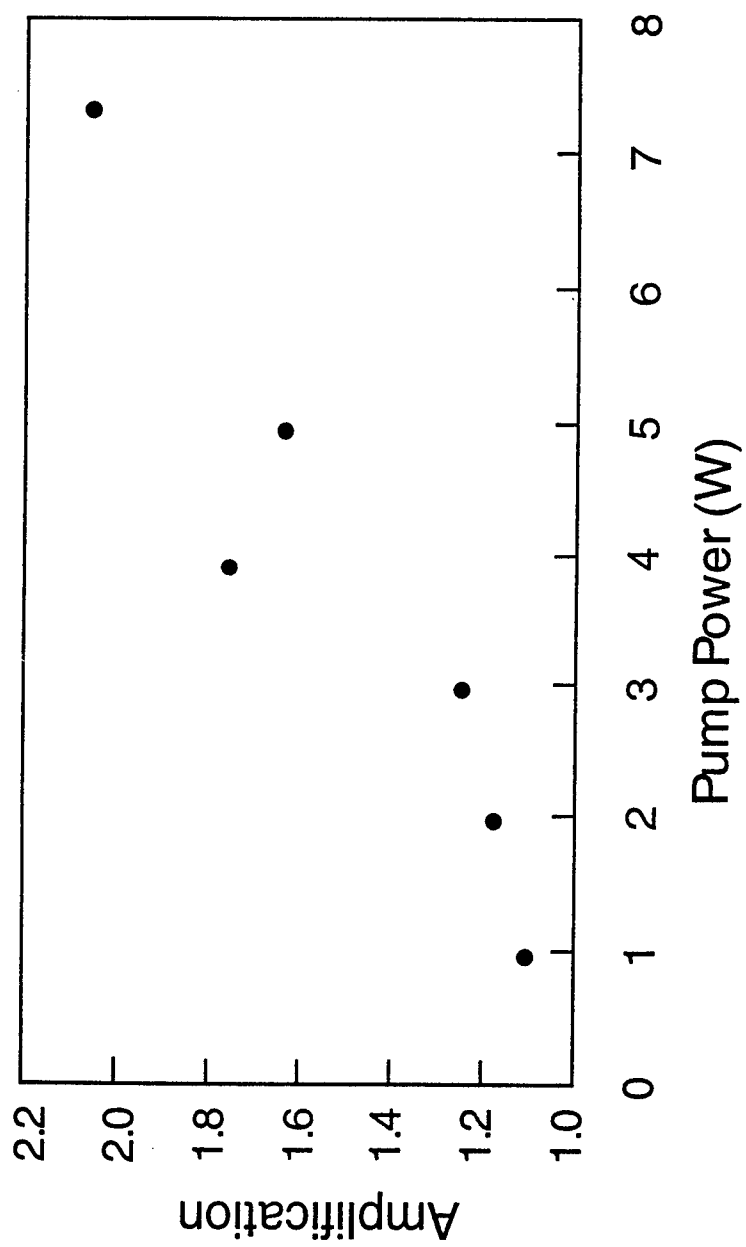


Fig.3. Net gain obtained at 1.23 μm from the Cr:forsterite composite amplifier

Material Science and Engineering Department on this issue.

We have had better luck with cleaving the polymer waveguides using elevated temperature. We found that heating the waveguide/substrate to 80° C, scribing the glass, and then applying a small amount of water to the scribe leads to cleaner fractures of the waveguide. Figure 4 shows a Scanning Electron Microscope picture of a cleave. The figure shows the top view, looking down on the substrate. The cleave is still not a well-defined straightline, but instead shows steps and structure with dimensions on the order of 50 nm. Within a region of 20 nm, the surface looks reasonably flat, so we hope this will allow reduced coupling loss. We are in the process of exploring various cutting edges, scribes, and glass thicknesses. One result we have obtained concerns post-processing of a cleaved waveguide. If the ragged-edged polymer cut is annealed for two hours at 200° C, the polymer film retracts from the edge of the glass and smooths out. The distance from the edge of the substrate and the edge of the smooth polymer waveguide ranges from 10-100 μm . We are investigating the use of index matching fluids and glass cover slips as one way of optically contacting the waveguide to the edge of the substrate.

We have recently extended this result by exploring the gain in a composite waveguide made using Cr-doped diopside. Diopside is related to forsterite, with a chemical structure of $\text{CaMgSi}_2\text{O}_6$. Like forsterite, diopside has both tetrahedral and octahedral sites for the Cr to reside. This implies that the Cr can be incorporated as either Cr^{3+} or Cr^{4+} . The Cr^{4+} is the preferred ion for infrared emission. Our first results with Cr:diopside show that the majority of the Cr is incorporated as Cr^{3+} . This has a large emission near 980 nm when pumped by an Ar-ion laser at 514 nm. Initial experiments with gain at 980 nm have been inconclusive. Using an experimental set-up similar to the one shown in Fig. 3, we have pumped a diopside composite waveguide with an Ar-ion laser at 514 nm, and probed the gain with a tunable Ti:sapphire laser at 980 nm. We observe a weak gain of approximately 7% in a 1 cm path length, as shown in Fig. 5. This result is significant because optical gain has never been reported in diopside before, simply because no one has succeeded in growing large single crystals of doped diopside. The diopside result illustrates the novel characteristics of the optical composite material.

IV. Femtosecond Pulse Generation using Cr:YAG Lasers

One of the major applications of the Cr:YAG and Cr:forsterite lasers is in optical communication. Most forms of optical communication today are done with Time Domain Modulation (TDM), where information is sent in the form of optical pulses which represent marks or spaces. The Cr:YAG and Cr:forsterite lasers are capable of operating with femtosecond pulses of high intensity, which enables the study of ultrafast switching phenomena in semiconductor devices, and in the exploration of nonlinear propagation in glass optical waveguides. Several research groups around the world have reported successful modelocking of these lasers with femtosecond pulses. The reported modelocking scheme relies on Kerr Lens Modelocking (KLM) to achieve the ultrashort pulses. We have discovered that there is a vast difference between simply obtaining short femtosecond pulses, and in getting femtosecond pulses that can be used in an experiment. Most KLM results require extremely sensitive adjustment of the laser, which requires hours of tedious mirror alignment. Once achieved the modelocking is not stable against most perturbations, and

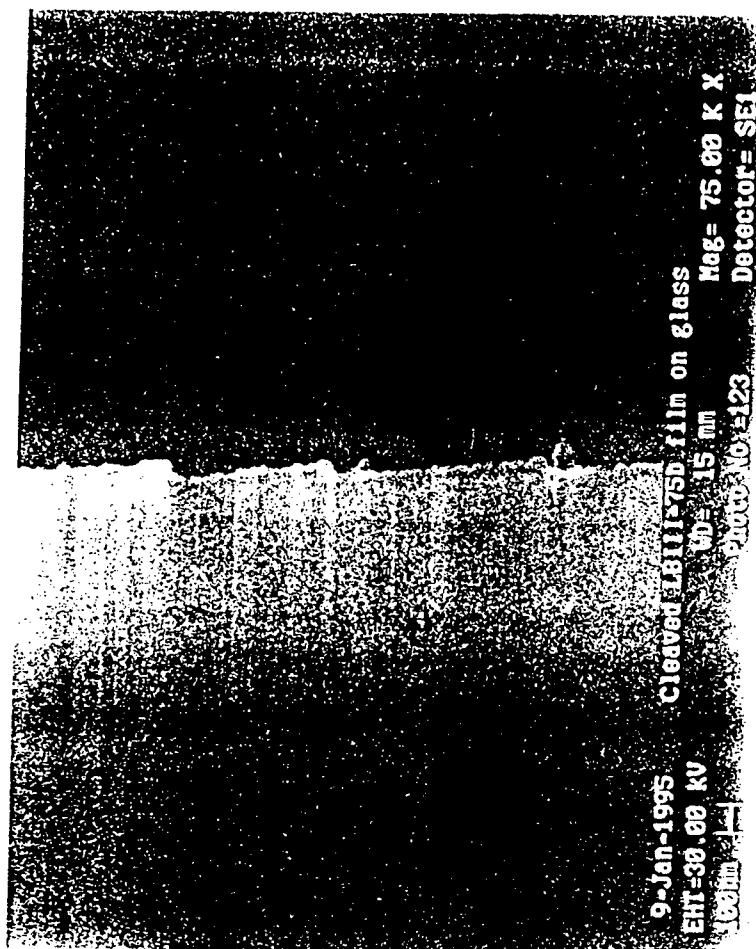


Fig. 4. SEM photo of a cleaved edge of the polymer-coated substrate.

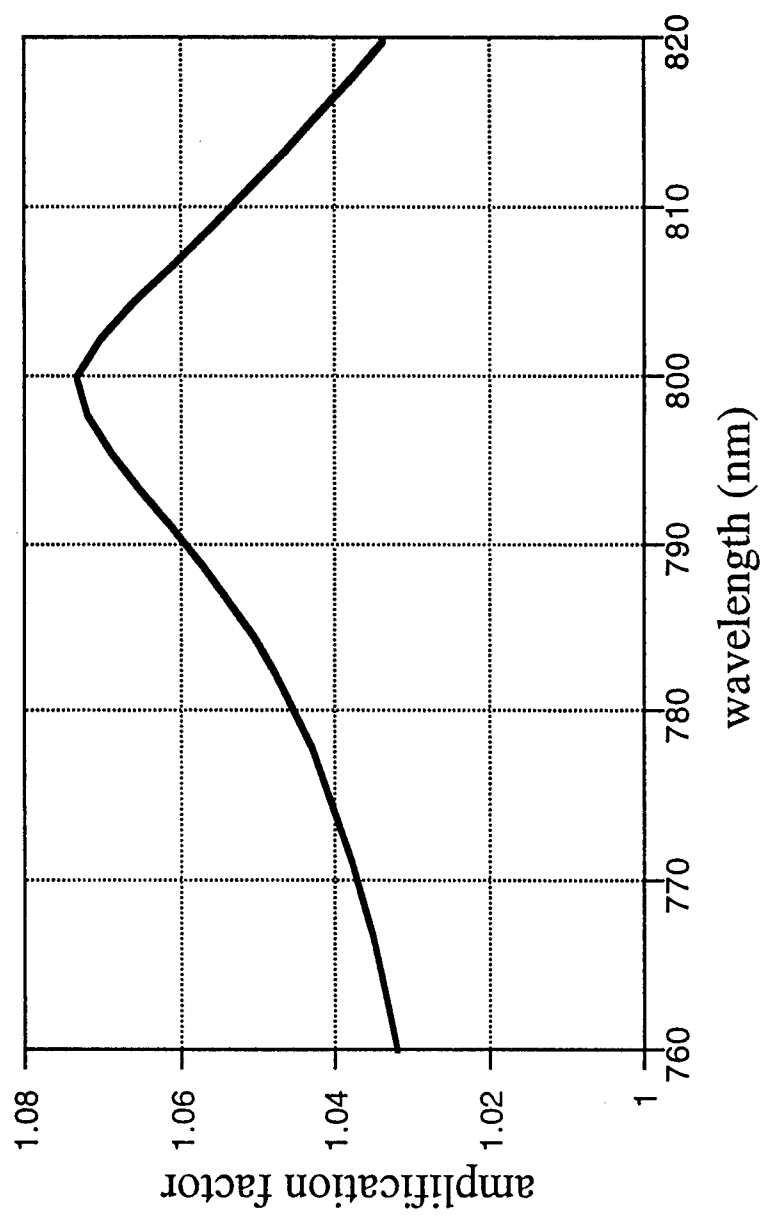


Fig. 5. Gain from a Cr:diopside composite measured with a tunable Ti:sapphire laser.

hence it has been very difficult to apply these femtosecond sources to the measurement of any physical system. Over the past year we spent a great deal of time attempting to stably modelock the Cr:YAG and Cr:forsterite lasers, without success. At best we could achieve moments of femtosecond output from either the Cr:YAG or the Cr:forsterite laser.

We continued to work on this problem in close collaboration with the personnel at Rome Laboratories. Lasers were set up both at Rome Labs and at Cornell, with a goal of achieving a stable train of modelocked pulses for experimental use. Initial efforts at Cornell concentrated on Cr:forsterite, while the Rome Labs effort focussed on Cr:YAG. At Cornell, graduate student Martin Jaspin duplicated the experimental set-up that Alphan Sennaroglu had used two years prior to successfully modelock Cr:forsterite. With exceptional care, he was able to achieve brief bursts of femtosecond pulse generation, but never was the output sufficiently stable to take experimental data. The problem is thought to be due to strong thermal lensing in the crystal. The transmitted pump beam shows significant distortion due to thermal blooming as the pump power is increased to several watts. This thermal lens is probably overwhelming any Kerr lensing that is occurring due to modelocking. With very careful alignment and stable pumping, it is possible to adjust the cavity in such a way as to compensate for the thermal lensing. However, this adjustment has large hysteresis, and is quite difficult to achieve in practice. Martin Jaspin had only limited success with this laser.

A talk at the Advanced Solid State Laser Conference[5] in San Francisco (January 31, 1996) demonstrated a simple method for modelocking the Cr:YAG laser using a Saturable Bragg Reflector (SBR). The basic idea had been demonstrated in June 1995 using a saturable Bragg reflector in a tunable Ti:sapphire laser. The basic idea is to replace one of the end mirrors of the laser with a semiconductor device which consists of a reflecting mirror (the Bragg reflector) and a saturable absorber. In the case of our Cr:YAG laser, the Bragg reflector consisted of 24.5 periods of a GaAs/AlAs quarter wave stack, and the saturable absorber consisted of two quantum wells tuned to have an exciton absorption near $1.5 \mu\text{m}$. We used a double quantum well structure consisting of 70 Å Ga_{0.47}In_{0.53}As / 80 Å Al_{0.48}In_{0.52}As / 70 Å Ga_{0.47}In_{0.53}As embedded in an Al_{0.48}In_{0.52}As quarter wave layer. Details of the construction of device reported at the Advanced Solid State Laser Conference were not made available, so we estimated construction in consultation with Rome Laboratories. Rome Laboratories had an SBR grown for the Cr:YAG laser by Dr. Rich Levitt at Adelphi. A second Saturable Bragg Reflector was designed to operate at $1.23 \mu\text{m}$ for the Cr:forsterite laser, and was grown by Dr. Bill Schaff of Cornell.

The performance of the SBR in the Cr:YAG laser has been truly impressive. The Rome Laboratories laser produced femtosecond pulses almost immediately upon its insertion into the laser cavity. The laser is extremely reliable and has controllable tuning. Significantly, the tuning range of the device spans from 1488 nm to 1535 nm. This is significantly greater than reported by the Bell Labs group. The use of saturable absorbers for modelocking has a broader tuning range than previously expected.

A sample of the SBR was brought to Cornell and used in a NaCl color center laser which operates over the 1.45-1.8 μm band. With one days solid effort we were able to achieve stable femtosecond pulses from the color center laser. The fact that the NaCl laser is extremely high gain, while the Cr:YAG laser is extremely low gain, demonstrates the

robustness of the SBR technique for generating femtosecond pulses. The SBR modelocking scheme appears to be very robust, and will certainly change the way we try to generate femtosecond pulses in the future.

A characteristic which is unique to the SBR modelocking is that the output power of the modelocked laser is limited to the 20-80 mW range. Using KLM modelocking, we have never observed an upper limit to the power. But with the SBR, there is definitely an upper limit to operational power before the femtosecond operation ceases. Best results are obtained at lower power, with typical outputs near 50 mW. This is true with both the Cr:YAG laser and with the color center laser. The reason for this is not understood at this time.

We were not able to modelock the Cr:forsterite laser with the SBR device grown here for that purpose. After characterizing the absorption features in the 1.23 μm SBR, we found that it did not display the absorption or saturation features we were expecting. In consultation with personnel at Rome Labs, we discovered that our sample was grown at low temperature as a way to avoid excess strain due to lattice mismatch. Unfortunately, low temperature growth with the quaternary materials systems leads to poorly defined boundaries and excess surface roughness. The quantum wells probably did not form, hence the device failed to provide the saturable behavior necessary to implement modelocking of the Cr:forsterite laser.

V. Conclusion

In this work, we have demonstrated the operation of a novel new optical material based on optical composites. Optical composites have many potential advantages: 1) they can be formed into optical waveguides, allowing long interaction lengths; 2) using copolymers, it should be possible to achieve adequate refractive index matching so that Fresnel scattering losses are insignificant compared to gain; and 3) in view of the high differential gain achieved in the waveguide, certain optical properties of nanocrystals may be superior to those of bulk crystals (e.g. increased dopant concentration, lower stress, etc.). Our result of 3 dB/cm at 1.25 μm demonstrates that the concept works. We now need to address the technical issues of end face preparation and cleaving.

The results with the Saturable Bragg Reflectors has been most impressive. Collectively researchers at Rome Laboratories and at Cornell have tried for years to find a reliable method for modelocking these Cr-doped lasers. While we have had occasional success with Kerr Lens Modelocking and Regenerative modulation, we have found from bitter experience that these techniques are not reliable. The SBR provides the first truly reliable method for pulse generation which will certainly lead to the ultimate application of these lasers to time-domain measurements.

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